SUPERHEAVY NUCLEI

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SUMMARY

The article is a short overview over the history and the present status of transuranium and superheavy element research, but there are presented here also not yet published data and results of experiments. As an introduction the first physical assumptions and proposals and the first attempts to create nuclei beyond uranium are presented. The neutron capture and consequent beta-decays, used at Berkeley to build transuranium elements up to Z =101, is the content of the second part. The series of asymmetric hot fusion reaction of uranium and transuranium nuclei with light ions led to new tranuranium elements of Z = 102 –106. Further progress was enabled with more symmetric cold fusion reactions of Pb and Bi targets with heavy ions. At present the last element, synthesized in this way has an atomic number of Z = 112. The production cross-section of Z =112 is below one pb. This value at present poses as an experimental limit for reasonable duration of a fusion experiment. The decay chain of a detected Z = 112 nucleus is used to explain the α-α correlation method of single superheavy nucleus identification. Further progress was made with hot fusion type reactions of tranuranium targets bombarded with double magic ^{48}Ca accelerated ions. Few α-decay chains of elements of Z = 114, 116 and 118 were observed, but the experimental results have to be confirmed in an independent way. The last part of the article is devoted to a short presentation of theoretical models, trying to describe the properties of the heaviest nuclei and the process of their synthesis at the fusion of two nuclei in a nuclear reaction. At the end of the article the open questions of the physics of superheavy elements are presented: Which is the maximum possible number of protons and neutrons in an atomic nucleus? Which number of protons creates the next closed shell? How large will be the stability enhancement at a double magic superheavy nucleus? How long can leave the most stable superheavy nucleus? The possibility to answer these questions is considered at the end of the article.

Keywords: *transuranium, superheavy, nuclei, nuclear reaction, half-life, cross section, separator, cyclotron*

1. INTRODUCTION

The first physical hypothesis trying to explain the atomic structure was presented by J. J. Thompson more than hundred years ago (1898) shortly after the discovery of the electron. The first experimental investigation of the atomic structure was accomplished by Geiger and Marsden in 1919 under the guidance of E. Rutherford. The results of the experiment led E. Rutherford to the formulation of the atomic model of a positively charged nucleus of the size of 10^{-15}m surrounded with negatively charged electrons. In the same year of 1919 E. Thompson successfully accomplished the first nuclear reaction when bombarding nitrogen with α-alpha particles observed the emission of positively charged particles – protons:

$$^{14}\mathrm{N} + {}^{4}\mathrm{He}\ (\alpha\text{-particle}) \rightarrow {}^{+}\mathrm{p}\ (\text{proton emission})$$

Enrico Fermi as soon as in 1934 supposed that the bombardment of nuclei of the heaviest known element – uranium with neutrons could create nuclei of elements heavier than uranium. The progress in the construction of particle accelerators, namely the cyclotron by Lawrence and Livingston in 1936, allowed verifying the assumption of Fermi about elements beyond uranium.

2. THE FIRST TRANSURANIUM ELEMENTS

The first transuranium element named as neptunium – Np was created in 1940 at Berkeley, California, by E. M. McMillan in a nuclear reaction

$$^{238}\mathrm{U} + \mathrm{n} \rightarrow {}^{239}\mathrm{U}\ (T_{1/2} = 23\ \mathrm{min}) + {}^{0}_{-1}e \rightarrow {}^{239}_{93}\mathrm{Np}$$

$$T_{1/2}\ ({}^{239}_{93}\mathrm{Np}) = 2.3\ \text{days}.$$

Measurable amount of $^{239}_{93}$Np, about 10 μg, was produced in 1994 in a nuclear reactor.

The next transuranium element of Z = 94 was observed at Berkeley in 1941 bombarding uranium with accelerated ions of deuterium

$$^{238}\mathrm{U} + {}^{2}_{1}\mathrm{H} \rightarrow {}^{238}\mathrm{Np} + 2\,{}^{1}_{0}\mathrm{n} \rightarrow {}^{238}_{94}\mathrm{Pu} + {}^{0}_{-1}e$$

Using the method of consequent neutron captures and beta decays heavier and heavier transuranium elements were created in the cyclotron laboratory at Berkeley. The method exhausted its capacity at the 101st element – mendelevium - $_{101}$Md synthesized in 1955 [1]. For further progress new experimental technique was developed – ion sources of intensive C, N, O and Ne ion beams and also cyclotrons, able to accelerate these ions to the desired energy.

Cyclotrons accelerating "heavy" ions were designed at Berkeley (California), at Kurchatov Institute (Moscow) and at the Joint Institute for Nuclear Research (Dubna).

At sharp competition of these institutes several transuranium elements were synthesized using suitable combination of transuranium target nuclei and bombarding ions:

1958: ${}^{244}_{96}\mathrm{Cm} + {}^{12}_{6}\mathrm{C} \rightarrow {}^{254}_{102}\mathrm{No} + 4\,{}^{1}_{0}\mathrm{n}$

1961: ${}^{243}_{95}\mathrm{Am} + {}^{16}_{8}\mathrm{O} \rightarrow {}^{256}_{103}\mathrm{Lr} + 5\,{}^{1}_{0}\mathrm{n}$

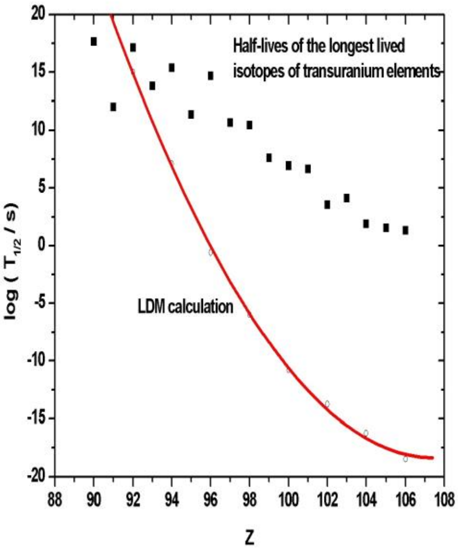
1964 - 69: ${}^{249}_{98}\mathrm{Cf} + {}^{12}_{6}\mathrm{C} \rightarrow {}^{257}_{104}\mathrm{Rf} + 4\,{}^{1}_{0}\mathrm{n}$

1970 -71: ${}^{249}_{98}\mathrm{Cf} + {}^{15}_{7}\mathrm{N} \rightarrow {}^{260}_{105}\mathrm{Du} + 4\,{}^{1}_{0}\mathrm{n}$

1974: ${}^{249}_{98}\mathrm{Cf} + {}^{18}_{8}\mathrm{O} \rightarrow {}^{263}_{106}\mathrm{Sg} + 4\,{}^{1}_{0}\mathrm{n}$

What we have learned from the investigation of transuranium elements:

a) The stability of nuclei of transuranium elements is orders of magnitude higher than the values given by the liquid drop model of atomic nuclei. The difference is increasing with increasing atomic number Z as it is shown in fig.1 where the half-lives of the longest-lived isotopes of transuranium elements is plotted as a function of Z.



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isotopes of transuranium elements compared with liquid drop model calculations

b) The longer half-lives due to the stabilizing effect of the nuclei shell structure. V. M. Strutinsky in 1967 [2] presented a method of quantitative calculation of the shell stabilization effect on heavy nuclei. The following calculations of half-lives of superheavy nuclei with closed shells were extremely encouraging, they were comparable with the half-lives of thorium and uranium. Consequently intensive search started to discover superheavy elements in terrestrial and extraterrestrial samples. Negative experimental results and improved shell effect and half-life calculations led to considerable shortening of the proposed superheavy half lives.

c) The production cross-section of tranuranium elements, synthesized in complete fusion reactions, decreases exponentially with increasing atomic number Z. This trend continues further to the heaviest known superheavy nuclei as shown in Fig. 2, where the production cross section of nuclei synthesized in cold fusion nuclear reaction, based on Pb and Bi target atoms and 1n deexcitation channel are presented.

3. THE COLD FUSION REACTIONS

When attempts to synthesize nuclei of elements with Z > 106 in hot fusion type reactions were repeatedly unsuccessful a new method of synthesis was proposed, the cold fusion reaction. In the previous hot fusion reactions the target nuclei were bombarded with ions of energies above the fusion barrier of the given target-ion system. In such reactions the fusion probability is high, but the formed compound nucleus is exited to energy of 30 –50 MeV and the probability of its prompt fission is adequately high. The surviving compound nuclei are emitting several neutrons to come to ground state energy level.

On the contrary, in the proposed cold fusion reactions more symmetric target-ion combinations, based on the double magic ${}^{208}\mathrm{Pb}$ or its neighbor

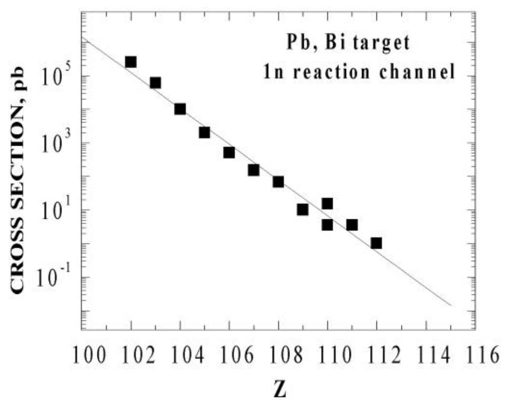

Fig. 2 Experimental cross-section values of transuranium and superheavy nuclei synthesized in cold fusion reaction using Pb and Bi targets.

^{209}Bi target nuclei, the system is fusing slightly below the fusion barrier and the compound nucleus is exited only to 10 – 20 MeV. The fusion probability is lower but the probability of prompt fission is considerably lower. The deexcitation goes through the emission of only one neutron.

The more symmetric target-ion combinations required a new accelerator, able to accelerate heavier ions at acceptable beam intensity and also a new kinematic separator, able to separate evaporation residues (ER) of higher energies than are produced in hot fusion reactions. Such an accelerator – separator system UNILAC-SHIP was put into operation in the new heavy ion research institute – GSI Darmstadt, Germany in 1979. Beyond few years three new heavy elements were synthesized in cold fusion reaction and after the upgrade of the SHIP parameters three more new elements were synthesized with atomic numbers Z = 110, 111 and 112 in years 1994–96. Because these nuclei are close to the supposed center of enhanced stability at Z = 114 they were designated as superheavy nuclei or superheavy elements:

<u>1981:</u> $^{209}_{83}Bi + ^{54}_{24}Cr \rightarrow ^{262}_{107}Bh + ^{1}_{0}n$

<u>1984:</u> $^{208}_{82}Pb + ^{58}_{26}Fe \rightarrow ^{265}_{108}Hs + ^{1}_{0}n$

<u>1982:</u> $^{209}_{83}Bi + ^{58}_{26}Fe \rightarrow ^{266}_{109}Mt + ^{1}_{0}n$

1990 – 94: 11 times increased sensitivity of SHIP:

<u>1994:</u> $^{208}_{82}Pb + ^{62}_{28}Ni \rightarrow ^{269}110 + ^{1}_{0}n$

<u>1994:</u> $^{209}_{83}Bi + ^{64}_{28}Ni \rightarrow ^{272}111 + ^{1}_{0}n$

<u>1996, 2002:</u> $^{208}_{82}Pb + ^{70}_{30}Zn \rightarrow ^{277}112 + ^{1}_{0}n$

Cross section $\sigma \approx 0.6$ pb (6×10^{-37} cm^2)

<u>Attempts to synthesize at GSI heavier nuclei in cold fusion reactions:</u>

<u>1995:</u> $^{208}_{82}Pb + ^{82}_{34}Se \rightarrow ^{290}116^* \rightarrow ^{290}116 + \gamma$

Cross-section limit $\sigma < 5$ pb (5×10^{-36} cm^2)

<u>1999 - 2000:</u>

$^{208}_{82}Pb + ^{86}_{36}Kr \rightarrow ^{294}118^* \rightarrow ^{283}118 + ^{1}_{0}n$

Cross-section limit $\sigma < 1$ pb (10^{-36} cm^2)

<u>Perspectives to synthesize at GSI new superheavy elements:</u>

The upgrade of SHIP increased the transport and detection efficiency of the system more than 11 times. It enabled at one picobarn cross-section level to register one decay chain of a superheavy nucleus in 10 days of continuous target irradiation. The present UNILAC-SHIP facility cannot be improved to the level, enabling to make experiments well below one picobarn level. To continue the successful cold fusion reaction method, based on Pb and Bi target nuclei below 1 pb, a new liner accelerator delivering high intensity not pulsed beam is required. The accelerator should be equipped with an adequately high current ECR ion source. Special problem is the effective cooling of the Pb target at such working conditions because of its low melting point.

4. METHODS OF IDENTIFICATION

An unambiguous identification of a new element requires determining the atomic and mass numbers of the registered nuclei. The first transuranium elements were identified by radiochemical separation methods. The production of only a very few nuclei of superheavy elements and their short half-lives exclude the radiochemical way of their identification. Almost all of the known superheavy nuclei are decaying preferably or exclusively by α-decay. This fact allowed the experimentalists to elaborate a very effective method of an unambiguous α-α correlation method of identification of new superheavy nuclei.

<u>The α-α correlation method:</u>

When an evaporation residue ER (synthesized superheavy nucleus) enter the detection system behind the separator it penetrates through two very thin carbon foils (about 30 μg/cm^2) of the Time-of-Flight (TOF) detector system which create two very fast signals. The ER ends its trajectory in a high resolution (14 –16 keV at 10 MeV) Si-detector. The impact of an ER is recorded with three signals – two fast TOF signals and a signal form the Si-detector.

The implanted ER after a sort time (from μs to ms, or seconds) decays emitting an α-particle. The α-particle creates a signal, which is not accompanied with TOF signals. The same implanted nucleus will successively undergo α-decays in time intervals in an agreement with the half-lives of the decaying nuclei of mass and atomic numbers according to the scheme

$$^{A}Z \rightarrow {}^{A-4}Z\text{-}2 \rightarrow {}^{A-8}Z\text{-}4 \rightarrow {}^{A-12}Z\text{-}6 \rightarrow {}^{A-16}Z\text{-}8 \rightarrow \ldots$$

In the case of cold fusion reactions the last members of the decay chain are nuclei of known half-life and α-particle energy, therefore they allow to identify the atomic and mass number of the nucleus emitting the first α-particle – the implanted ER.

There is one more very strong correlation factor, the position sensitivity of the Si-strip detectors. The Si-detector consists from many independent strips (at GSI 16 strips, each of them 5 mm wide and 35 mm high). The successively decaying nucleus emits α-particles from the same place of the same strip. The position resolution of the Si-strip detectors is about 0.1 – 0.2 mm what is enough for an unambiguous determination that all the recorded pulses of the decay chain originating from the successive decay of the same nucleus – the implanted ER. The pulse created at the implantation of the ER should come, naturally, also from the same position. One of the registered decay chains of element 112 is shown in fig. 3.

The situation is less favorable in the case when the implanted ER undergoes fission. There is there, in principle, the possibility of ER mass identification. The velocity v of the ER is measured with the TOF detector system and the kinetic energy E_k of the ER is measured with the Si-detector. According to the nonrelativistic energy-mass relation $m = 2E_k/v^2$ the mass of the implanted ER should be measurable.

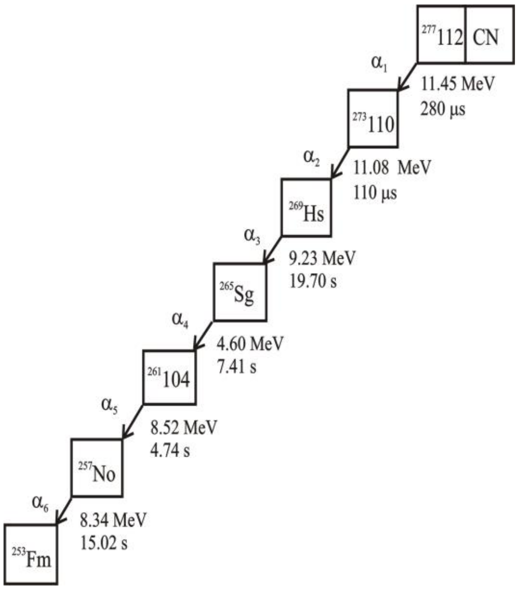

Fig. 3 The alpha-decay chain of the nucleus of element 112, synthesized in the reaction of ^{208}Pb(^{70}Zn,1n)^{277}112. Six α-particles α_1-α_6 were recorded in consequent time intervals shown in the figure. The measured α-particle energies are in agreement with values for the known members of the chain [3].

The main problem is the so-called plasma effect in the Si-detector, which causes in the case of energetic heavy ions nonlinear ionization and consequent pulse-height defect. The uncertainty of the pulse height defect excludes the possibility of ER mass determination.

5. THE RENAISSANCE OF HOT FUSION REACTIONS

The decrease of cross-section values of the cold fusion reactions, based on the double magic ^{208}Pb target nucleus below 1 pb at Z = 112 forced the physicists to examine other possibilities. As it was mention earlier, high excitation energies at more asymmetric hot fusion reactions led also to the problem of large decrease of cross-section values with raising atomic number Z. A promising proposal how to overcome the problem was given by Yu. Ts. Oganessian from the Laboratory of Nuclear Reactions, JINR Dubna, Russia. Oganessian proposed to use as bombarding ion the double magic $^{48}_{20}Ca_{28}$ calcium nucleus and as a target nucleus Pu, Am, Cm, Cf and Es transuranium element nuclei. The closed neutron and proton shells of ^{48}Ca should have a stabilizing effect on the fusion process. ^{48}Ca is a stable and the most neutron reach calcium isotope, but low abundant and therefore very expensive. The supplies of ^{48}Ca are only grams.

Attempts to synthesize new superheavy elements using ^{48}Ca accelerated ion beams are in progress at Dubna. Two types of kinematic separators, connected to the U400 cyclotron are in operation, but most of the experiments were made using the Dubna Gas Filled Separator – DGFS. The second kinematic separator – VASSILISSA is an ED-ED-ED-MD type ion optic system, completed with two triplets of quadrupole focusing magnets. After a series of test reactions experiments were made to synthesize superheavy nuclei of Z = 114, 116 and 118:

<u>1999:</u> ^{48}Ca + ^{244}Pu $\rightarrow$ ^{292}114* $\rightarrow$ ^{288}114 + 4n
Two α-decay chains were registered ending with SF of the granddaughter – ^{280}110.

<u>1999:</u> ^{48}Ca + ^{242}Pu $\rightarrow$ ^{290}114* $\rightarrow$ ^{287}114 + 3n
^{287}114 $\rightarrow$ ^{283}112 $\rightarrow$ SF
In this experiment two events were observed.

<u>2000:</u> ^{48}Ca + ^{248}Cm $\rightarrow$ ^{296}116* $\rightarrow$ ^{292}116 + 4n
One α-decay chain was registered ending also with SF of ^{280}110.

<u>2002</u>: ^{48}Ca + ^{249}Cf $\rightarrow$ ^{297}118* $\rightarrow$ ^{293}118 + 4n
This experiment is still running, one observed decay chain is tentatively assigned to the given reaction [4].

Tab.1 presents the list of all transuranium and superheavy elements, synthesized to the present days, or are under experimental investigation. The confirmation of the designation of elements 110, 111 and 112 by UIPAC is expected in short time

<u>The problem of unambiguous identification</u>: The decay chains of superheavy elements registered at Dubna in hot fusion reactions are less neutron

deficient than those, synthesized at GSI Darmstadt in cold fusion reactions. Therefore the position of the observed α-decay chains in the chart of nuclei is shifted to the right from the known area. For this reason the α-α correlation method of identification do not allow an unambiguous identification of the registered ER. There are, in principle, at least two possibilities to overcome this problem. The first possibility is to produce statistically significant number of events to proof the reproducibility of the observed experimental α-decay chains. But at picobarn and subpicobarn cross-section level it would need many years of beam-time. The second possibility is the direct measurement of the mass of the synthesized superheavy nucleus. One of the possibilities how to do that is in the process of realization at Dubna. The method, proposed by Yu. Ts. Oganessian [5] is based on the volatility of superheavy elements. The synthesized ERs after leaving the target are cached on the surface of

Tab. 1 Transuranium and superheavy elements

Z	Sym bol	Designation	Year of discovery	Place of discovery
93	Np	Neptunium	1940	Berkeley
94	Pu	Plutonium	1941	Berkeley
95	Am	Americium	1945	Berkeley
96	Cm	Curium	1944	Berkeley
97	Bk	Berkelium	1949	Berkeley
98	Cf	Californium	1950	Berkeley
99	Es	Einsteinium	1952	Berkeley
100	Fm	Fermium	1952	Berkeley
101	Md	Mendelevium	1955	Berkeley
102	No	Nobelium	1958	Berkeley Dubna
103	Lr	Lawrencium	1961	Berkeley
104	Rf	Rutherfordium	1964 1969	Dubna Berkeley
105	Db	Dubnium	1970 1971	Berkeley Dubna
106	Sg	Seaborgium	1974	Berkeley Dubna
107	Bh	Bohrium	1981	Darmstadt
108	Hs	Hassium	1984	Darmstadt
109	Mt	Meitnerium	1982	Darmstadt
110	*	-	1994	Darmstadt
111	*	-	1994	Darmstadt
112	*	-	1996	Darmstadt
114	**	?	1999-00	Dubna
116	**	?	2000-01	Dubna
118	***	?	2002	Dubna

* Naming is in progress at UIPAC
** Need additional confirmation
*** The experiment is not yet finished

a very hot metal catcher where from they immediately evaporate and are transported as low energetic ions to an ECR ion source. The ion source accelerates the ions to a uniform energy of 40 keV. The accelerated ions enter a mass spectrometer, which parameters are chosen for this special purpose. Behind the separator more than 200 Si-detector strips cover a wide sensitive area in the focal plane of the separator. The facility named as MASHA (Mass Analyzer of SuperHeavy Atoms) will have a mass resolution of about 0.3 atomic mass units and should enable safely determine the mass numbers of superheavy nuclei. MASHA is under construction, the first experiments are expected in 2003.

Some of the properties of MASHA will be known precisely enough only after a series of test experiments. Under question is the overall transport efficiency of the system, especially of the catcher and ECR ion source. The very low energy of the transported ions (40 keV) do not allow using a TOF detector, therefore there is no time signal. The impact of the very low energetic ion on the surface of one of the Si-detector strips is also without a response. Only those α-particles of the decay chain will be registered which are emitted in the direction of the Si-detector (50%).

7. THEORETICAL APPROACHES TO SUPERHEAVY ELEMENTS

The macroscopic properties of atomic nuclei were described reasonably well within the liquid drop model (LDM), but even at the time of the formulation of the model it was known that nuclei have properties which can not be explained or quantified in the framework of the model. More advanced versions of LDM use shell correction parameters. The extend and scope of this article do not allow to go into details of particular theoretical approaches to the properties of heavy and superheavy nuclei. All theoretical models combines, in different ways and ratios, the macroscopic and microscopic (one particle, shell) properties of the heaviest nuclei. Regardless of particular successes of the theory, a satisfactory description of the process of synthesis of superheavy nuclei is beyond the possibilities of the present-day theoretical models. Many unsuccessful attempts to predict the cross-section of formation of superheavy nuclei in complete fusion reactions clearly show, that we actually do not know what is going on when the bombarding heavy ion reaches the surface of the target nucleus and which parameters of the fusing system to which measure influence the fusion process

The survival probability of the synthesized superheavy compound nuclei is better understood. We have learned much about the shell structure and its positive impact on nuclear stability, expressed in nuclear binding energies, as deduced from experimental nuclear masses. We have solid knowledge about deformation of the heaviest nuclei. Today we know, that deformed nuclei also can have increased stability. We are able, after enormous overestimations some thirty years ago, to make more realistic half-life predictions, at least for even-even superheavy nuclei. Recent theoretical calculations, based on the Nilsson-Strutinsky model, predict for the available target-projectile combinations half-lives, which are longer than the experimentally critical 1 μs and shorter than 1 as it can be seen in Fig.4, where some calculated half-life values are presented 5]. The values presented with these half-life calculations differ from measured ones mostly by a factor smaller than 10 and in some cases the agreement is even much better.

7. CONCLUSION

The physics of the heaviest nuclei should answer the following fundamental questions:

- What is the maximum possible number of protons and neutrons in an atomic nucleus?
- Which number of protons create the next closed shell: Z = 114, 118, 120, or 126?
- At closed proton (Z= 114?) and neutron (N = 184) shells how large will be the stability enhancement?
- How long can leave the most stable superheavy nucleus: microseconds, hours, or millions of years?
- How large will be the production rate of the most stable superheavy nuclei?

After more than 60 years of intensive transuranium and superheavy element research all these questions are still open. It can be better understood why it is so, having in mind the complexity of the problem.

The problem of modeling the properties and behavior of the heaviest nuclei is first of all a many body problem of a system of almost three hundred partly independent elements. These elements are rearranged in the process of fusion in a large-scale dynamical transformation of the macroscopic and microscopic properties of the fusing nuclei.

To improve our model images we need more experimental points at the upper end of the chart of nuclei, especially around Z = 114, where model calculations, based on the Nilsson-Strutinsky approach [2] predict a closed proton shell with strong shell correction energies and spherical shapes of nuclei. Our possibilities to test theoretical predictions experimentally in this area of nuclei are limited. We are limited first of all due to the available combinations of stable or long living target and projectile nuclei.

We need more neutron rich combinations of target and projectile nuclei. This can be achieved only with radioactive beams. Despite of great progress in secondary beam intensities we are today still several orders of magnitude below the level what we need. In the case of Pb and Bi based complete fusion reactions even the most neutron rich radioactive nuclei of Zn, Ga and Ge do not allow to get close to the supposed double magic nucleus of $^{298}114_{184}$.

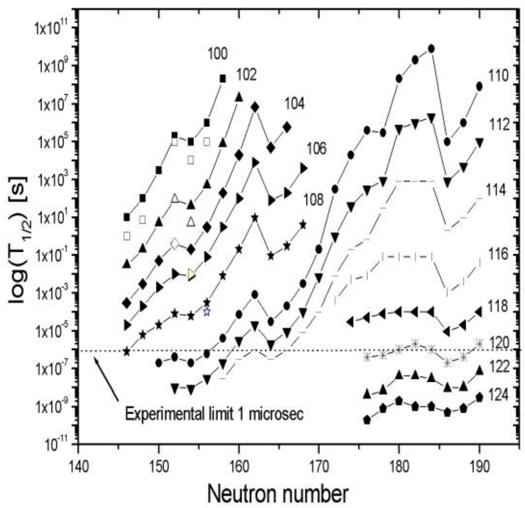

Fig. 4 Calculated α half-lives of nuclei of atomic numbers Z =100 – 124 and neutron numbers N = 140 – 190 [6].

The way on which we need to go further in our effort to answer the above listed questions is very difficult but not hopeless. The synthesis of transuranium and superheavy elements reached the border of experimental limits several times. As it was demonstrated in this article, these limits were always overcome. The ongoing scientific and technical progress will open new possibilities also in the field of the physics of superheavy nuclei.

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BIOGRAPHY

Štefan Šáro (1933) graduated as a physicist in 1957 at the Faculty of Natural Science, Comenius

University Bratislava. In the first period of his scientific carrier (1957-1985) main part of his research activity was oriented on low level counting and on application of the developed methods in applied and fundamental research. From 1985 his research activity is fully concentrated on the physics of superheavy elements. He is a co-discoverer of several superheavy elements. At present he is in the position of a professor of nuclear physics at the Faculty of Mathematics, Physics and Informatics, Comenius University, Bratislava.